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***In situ* measurement of elastic and total strains during ambient and high temperature deformation of a polygranular graphite**

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Abstract

In situ neutron diffraction and synchrotron X-ray diffraction, combined with image correlation analysis of 2D optical and 3D X-ray tomography datasets, have been used to investigate the relationship between elastic lattice strain and total strain during deformation of Gilsocarbon (IM1-24) polygranular nuclear grade graphite. The specimens were flat-end Brazilian discs under diametral loading, such that a compressive-tensile biaxial stress state was developed in the central region. The X-ray study was at ambient temperature, and the neutron diffraction was conducted at temperatures from ambient to 850°C. When under compression, there is a temperature-insensitive linear relationship between the total strain and the lattice strain that is measured perpendicular to the graphite basal planes. However, when under tensile stress, the total strain and elastic strain relationship is temperature sensitive: below 600°C, the lattice tensile strain saturates with increasing total tensile strain; above 600°C, significantly higher tensile lattice strains are sustained. The saturation in tensile lattice strain is attributed to microcracking in the graphite microstructure. Improved resistance to microcracking and damage tolerance at elevated temperature explains the increase in tensile strength of polygranular graphite.

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Keywords: Gilsocarbon graphite; neutron diffraction; X-ray diffraction; X-ray computed tomography, digital image correlation; digital volume correlation.

1 Introduction

Nuclear graphite is an artificial polygranular material with broad applications in nuclear energy due to its thermal and neutronic properties. It was used to construct critical structural core components (e.g. neutron moderator, fuel element, reflectors and core support posts¹) in the earliest reactors (e.g. the very first artificial nuclear reactor Chicago Pile-1 and the subsequent Clinton Pile) and it is a critical material in some operating reactors (e.g. the UK Advanced Gas Cooled reactors (AGRs) and Russian water-cooled RBMK reactors); a typical graphite moderated reactor may contain 2000 tonnes of graphite^{2,3}. Nuclear graphite is also a key material for some future Generation IV high temperature gas-cooled reactor (HTGR) designs; prototype HTGRs have been built in China⁴ (HTR-10 and HTR-PM) by licensing the German AVR design⁵, and the Japanese HTTR⁶, which has an outlet temperature of 950°C, has operated since the mid-1990's. The future potential for HTGRs is also demonstrated by ongoing national (e.g. India⁷) and international programmes (e.g. GEMINI and GEMINI+ between EU and USA), with the latter focussed on the design and regulatory framework of the first modern HTGR⁸. Reliable structural integrity assessment of graphite components is critical for new build designs^{9,10} and also for the safe operation and eventual decommissioning of the graphite cores in the current and retiring fleets¹¹. This must be underpinned by a sound understanding of the deformation and fracture of graphite under conditions that represent its service in the reactor environment of a high temperature gaseous coolant and fast neutron irradiation^{12,13,14,15}.

The structural integrity of graphite components is generally assessed using a maximum tensile stress criterion, such that failure is predicted to occur when the maximum principal stress exceeds an appropriate value. This is known to be simplistic and not fully satisfactory^{16,17,18} and other approaches, such as strain energy density, have been proposed¹⁹. However, a major difficulty with applying or validating any fracture criterion, particularly at stress concentrations such as notches, is that one needs to know the distribution of both stresses and strains. These are not linearly related for any kind of material behaviour that includes damage; polygranular graphite has non-linear and inelastic properties, particularly in

the un-irradiated condition and also as it approaches failure^{20,21,22}. To be able to reliably predict the structural integrity of graphite components, it is therefore necessary to have a detailed understanding of how stress causes strain, and how damage is accommodated by the graphite microstructure.

One of the most extensively studied polygranular graphite grades is Gilsocarbon (IMI-24). This was used to construct the UK AGRs and is typical of other medium/coarse-grained isotropic artificial graphites. Direct observations have demonstrated that its tensile strength is strongly related to pre-existing stress-concentrating defects in its microstructure^{23,24}. At ambient room temperature, *in situ* neutron and X-ray strain measurements have shown that for unirradiated Gilsocarbon in compression there is a linear relation between the elastic strains in the graphite crystals and the applied total strain, whereas in tension a non-linear behaviour occurs that is accompanied by a reduction in tensile elastic modulus and a relaxation of internal (i.e. thermally induced) stresses²⁵. This behaviour has been attributed to tensile microcracking, and studies of a propagating macroscopic crack in Gilsocarbon have also detected a fracture process zone^{26,27}, the effects of which could be described by a non-linear damage model²⁸. Similar modelling (traction separation laws based on maximum principal stress damage initiation and energy-based damage evolution with linear softening) has also been done for the modern fine-grained IG110 graphite^{29,30}. Synchrotron X-ray diffraction and imaging have both been applied to map the crystal strains and total deformation *in situ* within the fracture process zone of a propagating crack in unirradiated Gilsocarbon³¹. By using the experimental data for the effect of tensile strain on elastic modulus to calibrate the material damage model, a very good agreement was found between the predicted and measured crack stress fields. This showed that the non-linear deformation significantly reduced the stresses in the fracture process zone, and hence also reduced the elastic strain energy release rate available for crack propagation. The same behavior is reasonably expected to occur at stress concentrating notches, although this has not yet been demonstrated. An understanding of graphite non-linear behavior is therefore critical to the structural integrity assessment of graphite components.

The majority of studies of graphite deformation and fracture have been conducted at ambient room temperature, but graphite exhibits quite complex behaviour at elevated temperatures. For instance, the temperature dependence of the elastic modulus of unirradiated polygranular

graphite varies significantly with the test temperature, and also the graphite processing conditions that affect the graphite microstructure^{32,33}. At low temperatures, there is a negative temperature dependence of modulus that corresponds to the expected behaviour of its crystals but the temperature dependence of the elastic modulus can become positive at high temperatures (i.e. increasing stiffness with increasing temperature)^{34,35,36,37,38}. This has been attributed to a void-filling effect from thermally-induced closure of the Mrozovski accommodation pores. These lie between and within the graphite crystals, and form when cooling from graphitisation due to the crystal's highly anisotropic thermal expansion coefficients and elastic properties³⁹. The strength and fracture toughness of unirradiated polygranular graphite has also been reported to increase at elevated temperature and the crack propagation behaviour is also affected^{40,37,41}. Fast neutron irradiation further affects graphite behaviour in a complex manner through its influence on crystal dimensional change and other physical properties^{42,43}, but there are no data in the open literature for the elevated temperature mechanical properties of irradiated graphite.

To better understand how the graphite microstructure responds to stress and strain, *in situ* tests using neutron or X-ray diffraction can provide a “bridge” between the nano-scale characteristics of graphite, i.e. lattice strains in its network structure of fine crystals, and the macro-scale deformation of its heterogeneous porous microstructure. Our previous studies (e.g. ²⁵) have investigated unirradiated graphite at ambient temperature under flexural and tensile loading. The present work sets out to quantify the relationships between elastic strains and total strains in an unirradiated graphite at ambient and high temperatures. The elastic strains are measured using diffraction, to measure the change due to stress of lattice parameter of the graphite crystals, and the total strains are measured by image correlation, applied to two-dimensional optical and three-dimensional X-ray microscopy. A novel diametral compression setup using high temperature digital image correlation together with neutron diffraction has been adopted to investigate the effects of temperature. Additional insights and a validation of the neutron diffraction test methodology have been obtained by synchrotron diffraction and digital volume correlation of high-resolution computed X-ray tomographs at ambient (room) temperature. The methodology is suitable for examination of the behaviour of irradiated graphites at elevated temperatures, and these will be investigated in future studies.

2 Experiment

2.1 Material and loading configuration

The material was nuclear grade (IMI-24) Gilsocarbon graphite from the same billet as previous studies by some of the authors of this article^{25,44}. This graphite, which is used in the UK (AGR) Advanced Gas-cooled Reactors, is an iso-moulded coarse-structured polygranular artificial graphite with a nominal density⁴⁵ of 1.81 g/cm³ and total porosity²¹ of ~20 vol.%. It has weakly anisotropic properties, with a typical flexural strength ~30 MPa⁴⁶, tensile strength ~20 MPa and elastic modulus (both dynamic and static) of ~ 11 GPa^{16,43}.

The test methodology is based on diametral compression or the ‘Brazilian disc’ configuration⁴⁷. This is a convenient geometry for stable damage nucleation and fracture in brittle materials, and has been proposed for the evaluation of irradiated graphite properties for Gen. IV nuclear reactors⁴⁸. The test comprises a disc shaped specimen with loading applied at two diametrically opposed positions to produce a biaxial stress state that is compressive in the loading direction and tensile orthogonally. Reliable measurements of tensile fracture strength in the disc compression test are possible if the specimen fails in tension at its centre. This requires a sufficient compressive strength and also that the loading is sufficiently distributed along the disc periphery⁴⁹, otherwise neither the maximum tensile strain nor stress are located at the centre of the specimen and crack initiation from locations other than the centre is a possibility. The contact stresses can be reduced by cutting diametrically opposed and parallel flat surfaces, and both 2D⁵⁰ and 3D⁵¹ simulations of such flat-end Brazilian discs predict that, for a linear elastic material, the maximum tensile stress and strain are located at the disc centre. The test specimens employed in this study were flat end discs (Figure 1: diameter, D , 20 mm; thickness, t , 10 mm; $2\alpha = 29^\circ$), which were fabricated by electrical discharge machining. The tensile stress (σ_{yy}) that develops at the centre of specimen from compressive loading parallel to the direction of the x -axis, applied to the flat ends, is obtained for linear elastic properties as^{23,52,53}:

$$\sigma_{yy} = k \frac{2P}{\pi Dt} \quad (1)$$

and k is a correction factor for the contact angle ($k = 0.9185$ at $2\alpha = 29^\circ$):

$$k = \frac{(2\cos^3\alpha + \cos\alpha + \frac{\sin\alpha}{\alpha})^2}{8(\cos\alpha + \frac{\sin\alpha}{\alpha})} \frac{\alpha}{\sin\alpha} \quad (2)$$

Models⁵⁰ for this geometry also predict that for a homogeneous material following continuum linear mechanics, the displacement change, Δw , between the two flat ends has a relationship with the elastic Young's modulus, E , and Poisson's ratio, ν :

$$\Delta w = \frac{2P}{\pi Et} \left[(1 - \nu) - \ln \left(1 + \frac{4}{\sin^2\alpha} \right) \right] \frac{\alpha}{\sin\alpha} \quad (3)$$

Figure 1: Schematic of the loading configuration for the diametral compression or Brazilian disc test, with flattened ends at the loading positions. The contact angle (2α), subtended by the anvil contact area with the centre of the specimen, is 29° .

2.2 Measurement techniques

2.2.1 Neutron diffraction

Time-of-flight (TOF) measurements on the neutron spallation source at the ISIS Rutherford Laboratory, UK, were used for diffraction measurements of the graphite crystal lattice. The experiment was undertaken at the ENGIN-X beamline⁵⁴, where a high flux neutron beam with narrow pulse width is available with a 50 m primary flight path (L_1) to the test specimen. The diffracted neutrons are received at two collimated detector banks that are 180° (2θ) apart at $\pm 90^\circ$ to the incident beam (Figure 2a). The detected neutrons are from within an instrumental gauge volume (IGV) defined by the beam slits and detector collimation. In this study, the IGV was located at the specimen centre and had dimensions of $4 \times 4 \times 4$ mm. The distance between the IGV and the detectors is the secondary neutron

flight length ($L_2=1.53$ m), and the detected neutron wavelength, λ , is related to the measured flight time, t :

$$\lambda = \frac{h}{m(L_1+L_2)} t \quad (4)$$

where h is Planck's constant; m is neutron mass. The TOF spectra are then transformed to diffraction spectra, where each peak corresponds to a family of $\{hkl\}$ lattice planes in crystals that are suitably oriented for diffraction. From Bragg's law, the spacing of the lattice plane, d_{hkl} , is obtained from the corresponding TOF, t_{hkl} :

$$d_{hkl} = \frac{h}{2\sin\theta m(L_1+L_2)} t_{hkl} \quad (5)$$

where θ is the half angle between the incident neutron beam and the scattered path (i.e. 45° in the current setup). The calibrations were performed with a standard CeO_2 powder sample.

Figure 2: (a) Schematic (top view) of the experimental setup for neutron diffraction with loading, furnace, optical camera for DIC and neutron detector banks; There were two specimen configurations used (see Table 1) (b) #1:horizontal with the circular surface facing upwards and (c) #2:vertical where the flat circular surface faces the optical camera; (d) a typical neutron diffraction spectrum obtained for Gilsocarbon graphite.

The test specimens were loaded by a horizontal 50 kN servo-hydraulic Instron rig, within an infrared lamp-heated furnace that is integrated to the loading frame⁵⁵. Quartz windows in the furnace allow optical imaging by a digital camera and provide pathways for the incident and diffracted neutrons. In the high temperature tests, a flow of Argon gas was maintained through the furnace to protect the test specimen from oxidation. A white paint (VHT Flame Proof Very High Temperature Paint) was lightly sprayed manually onto the observed surface

of each specimen to provide the speckle for digital image correlation. The painted specimens were pre-cured in a vacuum furnace (120°C for 30 min, cool for 30 min, re-heat to 200°C for 30 min, cool for 30 min, final cure at 315°C for 30 min), which ensured the speckle pattern was maintained during the elevated temperature tests. After the pre-cure, the specimens were stored in a desiccator until tested at ENGIN-X, to exclude the potential influence of water vapour.

Two specimen configurations were used. In configuration #1, which is illustrated in Figure 2b, the test specimen is horizontal. This configuration uses the ‘Bank 1’ detector to measure the lattice spacings for planes that have their normal parallel to the compressive load, so the strain ϵ_{xx}^{ND} can be calculated using the change in lattice spacing, whilst the opposing ‘Bank 2’ measures the orthogonal strain, ϵ_{yy}^{ND} . In configuration #2, shown in Figure 2c, the circular surface of the specimen faces the optical camera. In this configuration, Bank 1 is again used to measure the compressive lattice strain, ϵ_{xx}^{ND} , whereas Bank 2 measures the orthogonal lattice strain, ϵ_{zz}^{ND} , through the specimen thickness. In the subsequent analysis of the elastic strain changes with applied load/stresses, only ϵ_{xx}^{ND} and ϵ_{yy}^{ND} are considered. Four temperatures were studied with applied loading: ambient room temperature (RT), 400°C, 600°C and 850°C, with at least two specimens tested in each configuration. During each neutron diffraction measurement, the specimen was held at constant load (i.e. using load control of the servohydraulic rig) to avoid parasitic stresses from thermal expansion or contraction of the loading train. Each diffraction measurement point required approximately 20 minutes to reach a count current of 15 μ A, with measurements obtained typically at 15 to 20 loading steps per specimen. A typical diffraction spectrum is shown in Figure 2d, with the stronger diffraction peaks labelled.

Prior to the diffraction experiment, a specimen was loaded at room temperature in configuration #2 in order to calibrate the optical camera. The single camera for optical measurements by 2D-DIC observed one surface of the specimen (Figure 2a), and a two-camera 3D-DIC (GOM-Aramis) system was introduced to observe the opposing surface. As the specimen was loaded to fracture, images from each camera set-up were recorded at each loading step. These were subsequently analysed by digital image correlation (DIC) to calculate the in-plane (2D and 3D DIC) and out-of-plane displacements (3D-DIC only). In the diffraction experiments, the two-camera 3D-DIC system was removed, and only the

single optical camera was used for in situ observations of the test specimen and loading anvils. The camera used for 2D-DIC is a BASLER PILOT PIA2400-17GM equipped with the Soyn ICX625 CCD sensor. Images were acquired at a resolution of 2456 x 2058 pixels, with a variable frame acquisition rate (by trigger). To avoid black body radiation effects⁵⁶ for the high temperature measurements, a TECHSPEC® blue bandpass filter (wavelength: 470 nm; full width half maximum: 85 nm) was placed in front of the optical camera. The pixel size for 2D-DIC was 12.8 μm and during DIC analysis to measure the displacement field with the LaVision DaVis 8.4.0 software (Version 42133), a decreasing interrogation window size from 128×128 to 96×96 pixel was used with an overlap step size of 75%. A filtering and smoothing operation was performed using a second order polynomial fit. For the 3D-DIC, the interrogation windows were positioned on the moving anvil since the specimen surface did not exhibit suitable contrast for correlation. The analysis used the GOM-Aramis software, with a facet (i.e. subset) size of 150×150 pixel at an overlap step size of 15; the pixel size was approximately 17 μm .

Table 1: Summary of the strain components measured from the corresponding test configurations employed in neutron diffraction (ND), X-ray diffraction (XRD) and X-ray tomography (XCT). Compression was applied along the x-axis and tension generated in the y-axis for all samples.

Analysis method	Configuration	Elastic strain	Total strain
Neutron Diffraction (ND)	#1	$\varepsilon_{xx}^{ND}, \varepsilon_{yy}^{ND}$	Interpolated from Configuration #2
	#2	$\varepsilon_{xx}^{ND}, \varepsilon_{zz}^{ND}$	$\varepsilon_{xx}^{DIC}, \varepsilon_{yy}^{DIC}, \varepsilon_{xy}^{DIC}$
X-ray Diffraction (XRD) & X-ray Computed Tomography (XCT)	#3	$\varepsilon_{xx}^{XRD}, \varepsilon_{yy}^{XRD}, \varepsilon_{xy}^{XRD}$	$\varepsilon_{xx}^{DVC}, \varepsilon_{yy}^{DVC}, \varepsilon_{zz}^{DVC}$ $\varepsilon_{xy}^{DVC}, \varepsilon_{yz}^{DVC}, \varepsilon_{xz}^{DVC}$

To analyse the neutron diffraction data, single-peak fits were performed using the ENGIN-X OpenGenie software⁵⁷. The fitted peak was a Voigt profile (mixed Gaussian and Lorentzian peak) convoluted with a sharp-edged exponential on a linear background. The lattice spacing, d , was taken to be the centroid of the Gaussian part of the fitted function. The lattice spacing of the basal plane peak (0002) was used to calculate the lattice strain, ε^{ND} (in units of micro-strain, $\mu\varepsilon$), using equation (6):

$$\varepsilon^{ND} = \frac{d_i - d_0}{d_0} \times 10^6 \quad (6)$$

where d_i is the (0002) lattice spacing at a given load and d_0 is the reference lattice spacing measured in the pre-load condition.

2.2.2 X-ray computed tomography and diffraction

The *in situ* X-ray computed tomography (XCT) and X-ray diffraction (XRD) strain mapping experiment was undertaken on the JEEP beamline I12 at the UK Diamond Light Source⁵⁸. The set-up is shown schematically in Figure 3a. The specimen was loaded between compressive anvils on a screw-driven Shimadzu rig, mounted on the large turntable in the second experimental hutch (EH2). A monochromatic beam of energy 61.86 keV was used and radiographs were recorded using the PCO.EDGE 5.5 sCMOS camera (2560 × 2160 pixel) manufactured by PCO AG, with optics selected to image an area of the specimen of 20 × 17.0 mm (7.9 μm per pixel). The lens for the module 2 datasets used in hereafter was custom-made by Still Optics GmbH from radiation tolerant glass. It has a focal length of 30.49cm. The exposure time per radiograph was 0.05 seconds, and we used an acquisition rate of 100 frames per second. The Stand-of-Distance was 2.35m.

The XCT scans were reconstructed from 2500 projections acquired over a restricted range of 146.2°, which was due to the shadowing from the steel pillars of the loading rig. In this limited angle tomography setup, the use of conventional reconstruction algorithms introduces image noise and artefacts that could affect the reliability of the digital volume correlation analysis^{59,60,61}. The tomography datasets were therefore reconstructed with the Conjugate Gradient Least Squares (CGLS) method⁵⁸, which is a fast-converging iterative algorithm that is less sensitive to the number of angular views. This algorithm, which uses the gradient descent method with conjugate directions, converged in 50 iterations for all scans. Each 16-bit greyscale tomograph obtained was finally cropped to a Field of View (FOV) of 2501 × 2150 × 1171 voxel, which is equivalent to 19.8 × 17.0 × 9.1 mm.

X-ray diffraction patterns were recorded using a large-area (42 × 42 cm) 2D Thales Pixium RF4343 detector with 2880 × 2881 pixel (each pixel 148 × 148 μm) positioned at a distance of 2.46 m from the specimen and calibrated using a CeO₂ powder sample. Due to physical constraints on the beamline, the transmitted beam was centred in the lower half of the

detector at approximately 13 cm below its midpoint thus partial Debye rings were recorded with a diffraction angle (2θ) of 3.4° for the (0002) basal plane. Beam slits of 1×1 mm were used, and the exposure time was 4 seconds per diffraction point. Stage movements of the 3-axis positioning table were used to map an area of 15×15 mm, in steps of 0.5 mm, on the circular flat surface (Figure 3b). Including the time for stage movements, the total time to acquire each diffraction map was 140 minutes.

Observations using tomography and diffraction were obtained at ambient temperature, initially in the load-free state (a small preload of 11 N was applied to hold the specimen in position) and then at loads of 1500 N, 3000 N and 4500 N that were applied using the screw-driven loading rig under displacement control. Specimen failure, identified by a detectable softening, occurred at a peak load of 5800 N. A tomograph was obtained at 5800 N, but a diffraction map was not recorded at this load as specimen failure had occurred. The changeover between tomography and diffraction modes at each load step was achieved by movement of the detector stages; the positioning was controlled with a reproducibility of better than $10 \mu\text{m}$.

Figure 3: (a) The loading configuration (#3) for X-ray computed tomography (XCT) and X-ray diffraction (XRD); (b) the area mapped by XRD, the gauge volume (GV) and the step size; (c) variation of normal strain $\epsilon_n^{\text{XRD}}(\phi)$ as a function of azimuth angle at a typical region (data and best fit). The expected measurement error for single points by XRD is $\pm 12 \mu\text{e}^{25}$.

Subsequently, Digital Volume Correlation (DVC) analysis on the reconstructed XCT scans (16-bit datasets with no preliminary filter) was performed using LaVision DaVis 8.4.0 software (Version 42133) on a Lenovo P900 workstation equipped with Xeon processors and Error Correcting Memory (ECC). Each of the scans at higher load was compared with the pre-load (11 N) dataset to derive the 3D displacement field in the deformed volume. Specifically, a Fast Fourier Transform (FFT) correlation with a large cubic subset ($224 \times 224 \times 224$ voxel dimension) at 50% overlap was first used to determine the large deformations. This was followed by the application of (i) an algorithmic mask to remove all voxels with an intensity equal or below that of the air to eliminate the contrast variation within pores, and (ii) a geometric mask to exclude the borders of the specimen. Finally, a series of four Direct Correlations (DC) with decreasing peak search radius, from 16 to 2 pixels, voxel binning, from $4 \times 4 \times 4$ to none and increasing passes, from 1 to 4, were used with the last step containing four passes of a cubic subset ($88 \times 88 \times 88$ voxel) at 75% overlap. The required valid voxel was 75%, which means that any voxel which is more than 75% outside the geometric mask is disregarded. The initial DC step used a uniform (square) subset shape function while the three later stages used a Gaussian function. As such, the calculated displacement vectors were mapped on a 3D spatial grid of approximately $348 \times 348 \times 348 \mu\text{m}$. There is no specific patterning technique as the contrast of the microstructure is sufficient for correlation, with datasets processed in 16 bit depth, with a useable contrast range of approximately 18167. The datasets were cropped and converted to RAWW format 16-bit unsigned little-endian, but no pre-filtering was applied before the correlation. The Quantities-of-Interest (QOI) were displacements obtained through the correlation and the derived strains. For visualisation purposes, post-processing included a displacement vector filtering of 0.7, a universal outlier detection with a remove threshold of 2 at $5 \times 5 \times 5$ neighbours as well as a Gaussian smoothing of $3 \times 3 \times 3$, one pass and with a strength of 0.5. For the subsequent XRD/DVC cross-analysis, a subvolume was extracted from the original results and exported without post-processing. A filtering of 0.7 with outlier removal was performed in the matlab code, but without any smoothing.

The rigid body movement of the specimen between the datasets were determined relative to the centre of mass in DVC and they were found to be relatively small compared to the dimensions of the $1 \times 1 \text{ mm}$ collimated X-ray beam. For example, the downwards vertical movement in the x -direction of applied load was less than $100 \mu\text{m}$ each load step, with a total

movement of 370 μm at 5800 N; the horizontal movement in the y-direction perpendicular to the X-ray beam was less than $\pm 15 \mu\text{m}$ each step, with zero net displacement overall; the horizontal movement along the z-direction parallel to the X-ray beam was less than 3 μm per step, except for the final step at 5800 N where a displacement of 42 μm was observed. The rigid body rotations were separated from the vector field deformation with polar decomposition and were all less than 0.3° . All of these movements were removed from the final 3D displacement fields, which had a displacement measurement precision estimated to be 0.065 μm . The 3D strain field was then calculated from the gradients of the displacement field, with a strain measurement precision of 264 $\mu\epsilon$ estimated from the displacement precision according to the final DVC subset size.

Prior to the loading of the sample, a Rigid Body Movement (RBM) was completed in order to obtain the strain precision at a later stage. A tomography was acquired, the sample table was moved by 900 μm (113 voxels) in the z-direction without any load applied to the sample, and a second tomography at preload was recorded, without changing any parameter such as exposure, number of projections or sample to detector distance. A DVC analysis was performed between the two datasets, with the same correlation parameters as the processing of the loading phase. This allowed us to determine the strain precision in X, Y and Z respectively at 39.05, 28.14 and 105.91 $\mu\epsilon$, which is below the value estimated with the empirical calculations.

For the XRD data processing, a cross-correlation based method was adopted here for improved precision in strain measurement in graphite compared with the conventional peak-fitting approach[†]. Specifically, an ellipse was fitted to the partial (0002) Debye ring to identify its centre. The intensity and position data were then binned into 1° radial sectors (ϕ) to obtain a set of radial intensity profiles generated by azimuthal integration within each sector. A cross-correlation routine, XCORR²⁵, determined the shift of peak position in each azimuthal direction relative to the load-free (i.e. 11 N pre-load) reference, and hence the normal lattice strain (ϵ_n^{XRD} , following equation 6). Best estimates of the strain components

[†] For neutron diffraction data, which has a lower signal to noise ratio in comparison to synchrotron XRD data, there is no significant benefit in using the XCORR analysis instead of conventional peak-fitting²⁵

ε_{xx}^{XRD} , ε_{yy}^{XRD} and ε_{xy}^{XRD} at each azimuth angle, for the purpose of mapping their spatial variation, could then be extracted using a least-squares fit of Mohr's circle to the azimuthal variation of normal strain, $\varepsilon_n^{XRD}(\phi)$ (Figure 3c) using:

$$\varepsilon_n^{XRD}(\phi) = \frac{\varepsilon_{xx}^{XRD} + \varepsilon_{yy}^{XRD}}{2} + \frac{\varepsilon_{xx}^{XRD} - \varepsilon_{yy}^{XRD}}{2} \cos 2\phi + \varepsilon_{xy}^{XRD} \sin 2\phi \quad (7)$$

3 Results

3.1 Reference (0002) plane spacing and thermal expansion coefficient

At ambient temperature, with a preload that was less than 1% of the failure load, the average (0002) plane spacing measured by neutron diffraction was 3.3754 ± 0.0003 Å (~40 measurements, ± 1 sample standard deviation) and that measured by synchrotron X-ray diffraction was 3.3722 ± 0.0001 Å (961 measurements, ± 1 sample standard deviation). For reference, Nelson and Riley⁶² first measured the *c*-dimension of the hexagonal lattice at room temperature in natural Ceylon graphite using powder X-ray diffraction to be 6.694 kx (i.e. 6.707 Å using a conversion factor of 1.00202⁶³) from which the (0002) plane spacing is 3.354 Å. This is slightly smaller (0.3%) than the value measured by ND and XRD. A larger (0002) plane spacing is expected in this artificial Gilsocarbon graphite since it is affected by the degree of graphitisation⁶⁴. It can also be affected by the thermal strains introduced during its manufacture, which also have been observed to be relaxed by mechanical loading²⁵. The measured difference in (0002) plane spacing between synchrotron ND and XRD can be attributed to the manual positioning of the CeO₂ standard prior to the in situ experiments. For example, the 0.18% difference would result from a relative position error of 4.5 mm (i.e. ~half the specimen thickness) at the ~2.46 m distance between the sample and detector. This would have occurred if the CeO₂ standard was placed at the same location as the graphite specimen surface. Nonetheless, although the absolute lattice parameter values for the synchrotron experiments may be slightly in error, this has no influence on the calculated strain values.

To examine the thermal expansion behaviour of the (0002) planes, specimens were heated (11 in total) during the ND experiment, with a small preload (50 N) maintained using load control, to temperatures of 200°C, 400°C, 600°C and 850°C at which neutron spectra was collected. In one case when a specimen had been heated to 600°C, the temperature was then

decreased to 200°C to obtain measurements on cooling (Figure 4). No difference was observed between the data from Bank 1 and Bank 2, and also no significant difference between heating and cooling. The crystal coefficient of thermal expansion (CTE), α_c , was obtained by linear fit to the data (the outlier data from the measurement at 850°C were included):

$$\alpha_c = \frac{\Delta d}{d_0 \Delta T} \quad (8)$$

where Δd is the change of plane spacing with a temperature change of ΔT (K or °C), and d_0 is the reference (0002) plane spacing (3.3754 ± 0.0003 Å) measured at room temperature. The average CTE obtained was $28 \pm 2 \times 10^{-6} \text{ }^\circ\text{C}^{-1}$.

Figure 4: The (0002) lattice plane spacing measured by neutron diffraction at different temperatures from the two detector banks 1 (closed symbols) and 2 (open symbols) (see Figure 2), with heating and cooling.

3.2 Load-displacement curve and fracture strength

Single-camera 2D-DIC was used to measure the specimen surface deformation during the elevated temperature tests at the ND beamline. Out-of-plane displacements can introduce erroneous strains in single-camera DIC measurements⁶⁵. However, the measurements by 3D-DIC (two-camera) (Figure 5a) show that the maximum out-of-plane displacement is insignificant ($< 20 \text{ } \mu\text{m}$ at failure load) compared to the sample to camera distance.

Furthermore, the in-plane net displacements measured by the 2D-DIC and 3D-DIC methods are closely comparable, hence the single camera 2D-DIC displacement measurements are considered to be reliable and were used throughout the experiment.

Figure 5: (a) Out-of-plane movement (z -displacement) measured by 3D-DIC ($<20 \mu\text{m}$ at failure load) and the in-plane x -direction net compressive displacements measured independently by 2D and 3D-DIC; (b) Comparison between DIC and direct measurements of the x -direction net applied displacements between the anvils (Error bars are smaller than the size of the symbols). The applied cross-head displacement is included for comparison; (c) Applied load as a function of the net applied displacement for all specimens at temperatures from room temperature to 850°C ; (d) The effect of temperature on tensile strength, with examples of the fractured specimens (neutron experiment data are presented as yellow circles, with average as grey circles, and the X-ray experiment data at room temperature as a blue triangle).

During the ND experiments, optical images of either the flat specimen surface (Configuration #2, vertical specimen, Figure 2c) or the side of the specimen (Configuration #1, horizontal specimen, Figure 2b) were obtained at each loading step. The loading anvils were within the field of view in both configurations. The net displacement applied to the specimen was derived via two alternate approaches: (i) using the 2D-DIC analysis to calculate the net difference in the average x -direction displacements of a flat region of dimension $3 \times 5 \text{ mm}$ located on each of the two loading anvils; (2) measuring the change in distance between the

edges of the loading anvils manually with the ImageJ software[‡] (“Direct Method”). An example of the data obtained by these two methods is presented in Figure 5b. Both the DIC and direct measurements gave consistent and equivalent results. The uncertainty (sample standard deviation) in the net displacement from the DIC measurements was 20 μm , and repeat measurements on the same images by the direct method had a similar repeatability (~ 25 μm , sample standard deviation). For comparison, the change in applied cross-head displacement with load is also presented in Figure 5b, and this shows the significant effect of compliance in the loading train. In the subsequent analysis, for those samples where reliable DIC could not be performed (e.g. due to saturated image intensity on the anvils), the direct method was adopted to obtain the applied net displacement.

The relationship between the net displacement across the specimen and applied load is summarised for the different temperatures in Figure 5c. The load-displacement behaviour appears linear below approximately 3000 N, with higher fracture loads and displacements attained with increasing temperature. The examples of fractured specimens (Figure 5d) show that they failed along the diameter that was parallel to the applied load. This is perpendicular to the maximum tensile stress, as expected in a successful disc compression test. In the ND experiments, the mean maximum tensile stress at failure, calculated using equation (1), increased with temperature (Figure 5d) from 16.7 ± 1.2 MPa at room temperature to 20.3 ± 1.2 MPa at 850°C. The specimen that was tested on the synchrotron X-ray beamline at room temperature failed at a maximum tensile stress of 17.0 MPa, which is consistent with those measured by the ND setup.

The elastic modulus, E , may be estimated from the load, P , and net displacement change, Δw , using equation (3). This was done with the data from Figure 5c, using a linear secant in the region from between 10% to about 50-60% of the peak load and assuming Poisson’s ratio, $\nu = 0.22$. The results, presented in Figure 6a as a function of the ratio of the load to peak load for example specimens, show no obvious dependence on test temperature. The measurement error for the modulus is around 1% for each data point due to the uncertainty in displacement measurement. The data are quite scattered (Figure 6a), and there are some

[‡] Schneider, C.A., Rasband, W.S., Eliceiri, K.W. "NIH Image to ImageJ: 25 years of image analysis". *Nature Methods* 9, 671-675, 2012

significant variations for individual specimens as the load increases. A histogram of all the modulus data from Figure 6a is plotted in Figure 6b, and the distribution has an average value (arithmetic mean $\pm$ sample standard deviation) of 7.5 ± 3.0 GPa.

Figure 6: Elastic modulus calculated using Wang's method; a) plotted as a function of the load/peak load ratio for the linear part of the curve for example specimens at different temperatures; b) Histogram distribution of the elastic modulus and the averaged data (mean and sample standard deviation) for all specimens at all temperatures.

3.3 Measurement of total strains (DIC and DVC)

The total strains were measured by image correlation, applied to 2D optical images (DIC) and 3D tomographic images (DVC) of the loaded specimens, relative to the load-free reference. Examples of the surface strain distribution (x - y plane), calculated as the gradients of the surface displacement field, are shown in Figure 7 at 20% and 90% of the peak load for a specimen tested at 850°C. As the load increases, the surface strain, ϵ_{xx}^{DIC} , parallel to the applied load becomes increasingly compressive at the centre, with a corresponding tensile increase of the orthogonal surface strain, ϵ_{yy}^{DIC} . Example data for the DVC calculated 3D strain distribution at ~25% peak load (1500 N) are shown in Figure 8, in which central slices from the x - y plane (parallel to the specimen's circular surface) and y - z plane (along thickness direction) are presented. The x -direction compressive strains, ϵ_{xx}^{DVC} , show a similar pattern as the DIC surface strains (Figure 7a) with a high level of axial compression at the loading contacts (Figure 8a) and orthogonal tensile strain at the centre due to the Poisson effect. Both DIC and DVC strain maps show some heterogeneities in the strain distributions, which may be due to local effects of the coarse Gilsocarbon microstructure.

For the DVC data, the strain distribution across the y - z plane (thickness) is generally constant. In light of this, the DVC-measured strains (ϵ_{xx}^{DVC} , ϵ_{yy}^{DVC} and the shear strain ϵ_{xy}^{DVC}) were averaged across the thickness of the sample (i.e. in the z -direction) and are presented in Figure 9 using interpolated contours. The data in the first 100 μm along the z -direction near either surface of the specimen have been excluded because the local approach adopted in the DVC algorithm tends to give unreliable results at surfaces and edges. In Figure 7, the DIC-measured strain distribution close to the sample edges have similarly been excluded. Figure 9a shows how the averaged ϵ_{xx}^{DVC} distribution evolves with increasing load. The highest compressive strains are at regions close to the contact points. Figure 9b shows the increasing tensile strain that is generated due to the Poisson effect. The shear strain, ϵ_{xy}^{DVC} , (Figure 9c) is negligible at the location of the neutron diffraction measurements in the central region of the volume.

Figure 7: Example of the surface orthogonal strains, (a) ϵ_{xx}^{DIC} and (b) ϵ_{yy}^{DIC} , obtained by DIC at 850°C at (i) low load (1.2 kN, 20% peak load) and (ii) high load (5.5 kN, 90% peak load).

The ND lattice strain were measured in the central $4 \times 4 \times 4$ mm IGV region. The DIC surface strain (ϵ_{xx}^{DIC} and ϵ_{yy}^{DIC} , Figure 7) and DVC strain through the thickness (ϵ_{xx}^{DVC} and

ϵ_{yy}^{DVC} , Figure 9) have therefore been averaged over the same region. A box representing this $4 \times 4 \text{ mm}^2$ central area is marked in Figure 7b(ii) and Figure 9a(iii) for reference. These averaged strains are presented as a function of the applied compressive load in Figure 10. The DIC compressive total strains (ϵ_{xx}^{DIC}) at different temperatures are shown in Figure 10a, overlaid by the compressive total strains from the RT DVC data (ϵ_{xx}^{DVC}) in the equivalent region of the specimen. Data for the tensile strains (ϵ_{yy}^{DIC} and ϵ_{yy}^{DVC}) are presented similarly in Figure 10b. Good agreement is demonstrated between the room temperature DIC and DVC strain data as a function of the applied load. There is a general tendency for the magnitude of the surface strains to decrease with increasing temperature, particularly at higher loads.

Figure 8: Examples sections of the 3D strains: a) ϵ_{xx}^{DVC} b) ϵ_{yy}^{DVC} , measured by digital volume correlation of tomographs at room temperature at 1500 N (~25% of peak load). The data are presented at the i) central x-y plane and ii) central y-z plane (plane locations marked with white dotted lines). The slices of the strain data are superposed on central slices of the tomograph, with a colour and transparency that varies with the strain magnitude.

The total strains (ϵ_{xx}^{DIC} and ϵ_{yy}^{DIC}) presented in Figure 10 are those measured on samples tested in configuration #2 during the ND experiment, for which the flat circular surface faced the camera (Figure 2c). For those samples tested at configuration #1, the sample's flat circular surface was facing upwards (Figure 2b) and could not be imaged. Due to the consistency of the DIC strains measured in repeat samples in configuration #2, and the agreement at RT with

the DVC strain derived from XCT tomographs (Figure 10), the configuration #2 relationship between total strain and load has therefore been interpolated to obtain the applied total strains as a function of load for those samples that were tested in configuration #1 (Figure 10). The configuration #2 DIC data in Figure 10 were fitted, in two groups (group01: RT & 400°C; group 02: 600°C & 850°C) using polynomial regression, to define these relationships.

Figure 9: 2-Dimensional DVC strain maps in the x - y plane (a) ϵ_{xx}^{DVC} , (b) ϵ_{yy}^{DVC} and (c) ϵ_{xy}^{DVC} ; obtained by averaging of the strains through the thickness (z -direction) of the specimen tested at room temperature and observed at compressive loads of (i) 1500 N; ii) 3000 N; and iii) 4500 N. The dashed lines identify the centre point of the specimen.

Figure 10: The effect of load on the average total strains: (a) compressive total strain, ϵ_{xx}^{DIC} and (b) tensile total strain, ϵ_{yy}^{DIC} , in the 4×4 mm region at the centre of neutron diffraction specimens from room temperature (RT) to 850°C. The data “DVC_RT” are the strains ϵ_{xx}^{DVC} and ϵ_{yy}^{DVC} averaged within a $4 \times 4 \times 4$ mm volume at the centre of the X-ray tomographed

specimen, tested at room temperature. The uncertainty for average total DIC strain (sample standard deviation of ~200 measurements within a 4×4 mm area) is about $130 \mu\epsilon$ in all samples, which is similar to the symbol size. The uncertainty for average total DVC strain (sample standard deviation of ~1300 measurements within the $4 \times 4 \times 4$ mm volume) is less than $100 \mu\epsilon$.

3.4 Measurement of elastic lattice strains (X-ray and neutron diffraction)

Diffraction measures the elastic strains in the graphite crystal lattice for those grains that are suitably oriented for diffraction. These data report the strain in the crystal direction perpendicular to the (0002) basal planes (i.e. along the c -axis). Examples of the synchrotron X-ray diffraction strain maps (ϵ_{xx}^{XRD} , ϵ_{yy}^{XRD} and ϵ_{xy}^{XRD}) obtained at RT at three different loading conditions (1500 N, 3000 N and 4500 N) are shown in Figure 11. The elastic strains, ϵ_{xx}^{XRD} (Figure 11a) which are parallel to the direction of the applied load, are largest in compression at the left and right sides of the map (i.e. adjacent to the loading points). The largest tensile elastic strains, ϵ_{yy}^{XRD} (Figure 11b), which are orthogonal to the applied load, are observed in the centre of the specimen. The shear strains, ϵ_{xy}^{XRD} (Figure 11c) increased with applied load, with a pattern corresponding to the total shear strains (Figure 9c). These patterns of elastic lattice strain are consistent with the total strain distributions observed by both DIC (Figure 7) and DVC (Figure 9).

Different from the XRD maps, the neutron diffraction measures only one data point for the strains at the centre of the sample averaged over a $4 \times 4 \times 4$ mm IGV (as shown in the schematic in Figure 2a). One neutron measurement was undertaken at each load; the lattice strains in the IGV (compressive: ϵ_{xx}^{ND} and tensile: ϵ_{yy}^{ND}) were calculated relative to the load-free (50 N pre-load) condition and plotted as a function of applied load for all samples (Figure 12). Both the average compressive and tensile elastic strains, ϵ_{xx}^{ND} and ϵ_{yy}^{ND} , increase in magnitude with increasing load. The through-thickness strain, ϵ_{zz}^{ND} , which was measured in configuration #2 was negligible ($< XXX$). The compressive lattice strains, ϵ_{xx}^{ND} , increased with the applied load following a linear trend with no clear dependence on temperature. The tensile elastic strains, ϵ_{yy}^{ND} , have significant scatter compared with the compressive strain, ϵ_{xx}^{ND} . The magnitude of the tensile elastic strains, ϵ_{yy}^{ND} , tends to increase and then saturate at around $200 \mu\epsilon$ with increasing load at both room temperature and 400°C . However, at 600°C and above, the tensile elastic strains are higher and also tend to plateau with a maximum value of $\sim 600 \mu\epsilon$. For comparison, the XRD strains, ϵ_{xx}^{XRD} and ϵ_{yy}^{XRD} , have been averaged

over the central 4×4 mm, and have been plotted with the neutron diffraction results as a function of the applied load in Figure 12a and 12b. These ambient temperature XRD data are quite consistent with the RT neutron diffraction strains.

Figure 11: Mapping of the lattice strains (a) compressive strain (ϵ_{xx}^{XRD}), (b) tensile strain (ϵ_{yy}^{XRD}) and (c) shear strain (ϵ_{xy}^{XRD}) by XRD at room temperature with compressive loads of (i) 1500 N; ii) 3000 N; and iii) 4500 N. The dashed lines identify the centre point of the specimen.

Figure 12: Neutron diffraction (ND) data for (a) compressive lattice strain, ϵ_{xx}^{ND} , and (b) the tensile lattice strain, ϵ_{yy}^{ND} , in the $4 \times 4 \times 4$ mm IGV as a function of applied load from room temperature (RT) to 850°C. The XRD_RT data are obtained at room temperature by X-ray diffraction (ϵ_{xx}^{XRD} and ϵ_{yy}^{XRD}) and are the average of the measurements in the central 4×4 mm region. The measurement uncertainty for the ND data is $\pm 50 \mu\epsilon$ ⁵⁴ and the sample standard deviation for the XRD data is ± 15 to $50 \mu\epsilon$, which is of the order of the measurement error²⁵. Error bars are added to the XRD_RT data, but the symbols are larger than the error bars.

3.5 The relationship between total strain and elastic lattice strain

The measurements using DIC (surface strain) and DVC (3D strain field) provide the total strains, and these are now compared with the diffraction-measured elastic strains in the crystal lattice (obtained by either ND or XRD) acting in the same direction. For the X-ray experiment at RT, each of the elastic strain data points (Figure 11) has been obtained from diffraction by suitably oriented grains in a pencil shaped gauge volume as the X-ray is transmitted through the sample in its thickness direction, and the measurement of normal strain around the Debye rings (Figure 3c) allows the local directions of maximum principal strain to be determined. The DVC analysis of the XCT tomographs on the same sample can resolve the 3D strain field within the same gauge volume, and the DVC strains have been averaged through the thickness of the sample to permit a direct comparison between the two measurements. As such, the directions of the principal strains derived from the strain maps for the two methods (elastic strain from XRD and total strains from DVC) are presented in Figure 13. This shows that the principal strains are well aligned with the loading axes (x and y within the central 4×4 mm region).

Hence, for cross-comparison between all the different techniques, data for ε_{xx} and ε_{yy} from the centre of the specimen can be used as follows: i) DIC data averaged over the surface (4×4 mm); ii) ND data from within the IGV of $4 \times 4 \times 4$ mm; iii) XRD measurements as the average of the 4×4 mm region; and iv) DVC measurements averaged in the central 4×4 mm region and across the sample thickness.

The observed relationship between the elastic lattice strain and total strain at RT, in the x and y directions parallel and perpendicular to the applied load, is presented in Figure 14a(i) for this central region. The XRD measurements (from Figure 9 and Figure 11) and ND data (from Figure 10 and Figure 12) are presented together in Figure 14a(i); these are labelled as ‘current data’. In addition, previously reported data²⁵ from experiments on the same grade of graphite, obtained at room temperature using four point bending with ND at the ENGIN-X beamline and uniaxial tension with XRD at beamline I12 (Diamond Light Source) have been overlaid on the current data; and these are labelled as ‘past data’. The total strains in Figure 14a(i) have been acquired by a range of methods including DVC and DIC in the current work, and DVC and resistance strain gauges in past work, therefore, the x -axis is labelled as ‘total strain’ without specifying the actual method. Comparing the current and past data from room temperature experiments shows consistency in both the tensile and compressive quadrants.

Figure 13: Maps of the direction of maximum principal strain, measured at room temperature by a) X-ray diffraction (XRD) and b) digital volume correlation of tomographs (DVC) with compressive loads of (i) 1500 N; ii) 3000 N; and iii) 4500 N. The dashed lines identify the centre point of the specimen. Lower quality XRD data at the edges of the mapped region have

been censored, so the mapped regions are not identical in area. The 4×4 mm region at the specimen centre that corresponds to the ND IGV is indicated.

Figure 13 has shown that the directions of the principal strains vary with position in a similar pattern for both lattice and total strains. Damage models for brittle and quasi-brittle materials such as graphite use criteria of maximum principal tensile strain^{16,17,18}. It is therefore important to measure the maximum principal strains by resolving the strain tensor. Hence, Figure 14a(ii) presents the relationship between the principal strains measured in the point by point XRD map and DVC analysis at the same locations in the full volume of the specimen (i.e. $\sim 15 \times 15$ mm mapped region). This was obtained using a natural neighbour interpolation and nearest neighbour extrapolation of the DVC measurements (Figure 9) onto the same measurement grid as the XRD observations (Figure 11). Due to the large amount of data, the plot is coloured by the number density of data points; a higher density is shown for the tensile data in comparison to the compressive data as the same number of data points occur within a smaller range of strain. There is an essentially linear relationship between the compressive principal total strain and elastic principal strain, whereas in tension the elastic principal strain saturates with increasing applied principal strain. These trends are unaffected by the orientation of the principal strain axes relative to the applied loading; in the central 4×4 mm region the principal strain axes are well aligned with the applied load and so the relationship between the principal strains is well described by measurements in the neutron diffraction IGV. This analysis demonstrates that diffraction, combined with image-based deformation measurement, is an effective method to obtain the relationship between the principal elastic lattice strains and total principal strains. It also demonstrates, for this disc compression experimental configuration, that comparison of ND data from within the central IGV with surface DIC observations in the central 4×4 mm region provides a satisfactory description of the relation between the principal elastic and total strains.

Figure 14: Lattice strains (diffraction) as a function of the total strain (a) room temperature data for (i) strains in the x (tensile) and y (compressive) directions within the 4×4 mm central region from the current neutron and X-ray studies, compared with neutron and X-ray data from a past study ref [25] (flexure and tension); (ii) maximum (tensile) and minimum (compressive) principal strains over the 15×15 mm mapped region, and (b) neutron diffraction from room temperature to 850°C . [In a(ii), the colour shows the relative number density of data points]

The ND data obtained in this experiment from room temperature to 850°C are presented in Figure 14b. Here again, there is a linear relationship between the applied total compressive strain and elastic lattice strain, and this has no observable dependence on temperature. At room temperature and 400°C , the tensile elastic lattice strains only attain a low value of $\sim 200 \mu\epsilon$ that does not then increase as the applied tensile strain increases above $1000 \mu\epsilon$.

However, at high temperature (600°C and above), the tensile elastic lattice strain tends to plateau at a higher value of $\sim 600 \mu\epsilon$ as the applied tensile strain exceeds 2000 $\mu\epsilon$.

4 Discussion

The compressive elastic strains, measured by diffraction from the (0002) planes, increase in proportion with the compressive total strain (Figure 14a). However, the magnitude of the elastic strains is only $\sim 20\%$ of the applied total compressive strain. The discrepancy is attributed to both the macro-porosity and Mrozowski cracks⁶⁶ (Figure 15a-c). As pointed out by Liu and Cherns⁶⁷, Mrozowski cracks and termination porosity are present between the crystal platelets due to crystallography requirements in Gilsocarbon graphite. These Mrozowski cracks are also responsible for the sensitivity of the bulk elastic modulus of artificial polygranular graphites to their processing conditions^{34,35,36,37,38}.

This can be understood by considering a simplified case, illustrated in Figure 15: if a total uniaxial tensile deformation, Δ_T , is applied, the measured elastic lattice strain, Δ_d , should be the same as the overall deformation (Figure 15a). However, Gilsocarbon graphite has a porous structure with large pores and smaller defects (e.g. Mrozowski cracks), so the total displacement, Δ_T , is a function of both the displacement of the crystalline matrix, Δ_m , and that of the macro-pores and defects, Δ_P , (Figure 15b). (Note: the parallel lattice planes indicated in Figure 15 are idealised for clarity of explanation as, in reality, the orientation of the crystals will be random). There are also stiffer regions in Gilsocarbon graphite (Figure 15c) due to the strong elastic anisotropy of the graphite crystals and local textures in the ‘onion-like’ filler particles. Large circular pores are usually associated with these particles (Figure 15c). If look closer to the nano-scale, Mrozowski cracks are also present between the platelets within a grain to share the local displacement. Taking these multiple length-scale features into consideration, the applied displacement is distributed amongst the constituent phases of this ‘composite’ microstructure:

$$\Delta_T = f(\Delta_m, \Delta_P, \Delta_P') \quad (9)$$

where: Δ_m is the displacement of the matrix graphite material in general without differentiating between the binder and filler particles, Δ_P is the displacement of the macro-pores and defects (including those ring shaped pores located around the filler particles and also those more

spherical-shaped ones embedded in the binder phase); and $\Delta_{P'}$ is the displacement accommodated by the nano- and micro-pores (e.g, Mrozovski cracks and nano-pores caused by terminating planes due to crystallographic requirement⁶⁷) at the crystal level. The actual lattice strain due to which is measured by diffraction from the (0002) planes, is thus less than the total strain applied to the bulk volume (e.g., only about 20% of total compressive strain was passed to the crystallites for decreasing the basal plane d-spacing, Figure 14b).

Variations in the orientation of the graphite crystallites and accommodation porosity in the microstructure (such as between the filler and matrix), and also the distribution and connectivity of larger scale porosity⁶⁸ affect the bulk elastic modulus (Figure 15b). Larger scale defects within the microstructure, which may evolve or propagate in response to mechanical loading, will also affect the relationship between the applied strains and the elastic strains from the stress in the graphite crystals (Figure 15c). The effects of mechanical damage will be considered in further detail later in this discussion.

Figure 15: Schematic showing the loading/displacement bearing mechanism in cases when a displacement is applied to: (a) a lattice; (b) a volume of material containing weak phases (pores, defects, etc) embedded in the matrix; (c) a volume of material containing weak phases (pores, defects, etc) in the matrix, stronger phases with pores separating it from the matrix and lattice-scale cracks that form between basal planes. Note the schematic did not consider the realistic random distribution of the pores and it is an idealised structure along one direction only to elucidate the different factors contributing to the overall displacement.

For polygranular graphites, assuming an idealised fully dense random assembly of graphite crystals, the CTE at room temperature at any direction can be estimated according to the Bacon orientation functions⁶⁹. The average CTE measured perpendicular to the basal plane, (α_c), was $28 \pm 2 \times 10^{-6} \text{ } ^\circ\text{C}^{-1}$, which is consistent with the expected value of $28.3 \times 10^{-6} \text{ } ^\circ\text{C}^{-1}$

measured over the same temperature range in natural Ceylon graphite, parallel to [0001]⁶². The CTE parallel to the basal plane (α_a) was not measured in this work, as reliable fitting to the {11 $\bar{2}$ 0} ND peak was affected by its proximity to the {11 $\bar{2}$ 2} peak, but the literature value⁶² is $-1.5 \times 10^{-6} \text{ }^\circ\text{C}^{-1}$. Assuming an isotropic distribution of crystal orientations, the bulk CTE α_x is:

$$\alpha_x \approx \frac{\alpha_c + 2\alpha_a}{3} = \frac{28 - 2 \times 1.5}{3} = 8.3 \times 10^{-6} \text{ }^\circ\text{C}^{-1} \quad (10)$$

Various studies^{43,70,71} have reported the bulk CTE of Gilsocarbon graphite to be $\sim 7 \times 10^{-6} \text{ }^\circ\text{C}^{-1}$; this lower value in comparison with simple theory may be due to the effects of texture and accommodation porosity in the graphite microstructure⁴³.

The compressive elastic modulus, estimated using equation (3) from the applied load and net displacement (Figure 6a), was unaffected by temperature. The effect of load on the compressive elastic strains is also insensitive to temperature (Figure 14b). Both are consistent with previous observations that the elastic modulus of graphite is relatively insensitive to temperatures below 1000°C ^{40,37}. However, the average modulus of $7.5 \pm 3.0 \text{ GPa}$ (Figure 6b), is significantly lower than literature data for Gilsocarbon^{16,43}. This may be because the applied net displacements were measured at the loading anvils. As in many contact problems, the stress concentration at or close to the contact faces caused higher deformation or local damage in the material. As such, the total net displacement used in equation (3) derived from the total displacement between anvils is larger than that in an ideally elastic condition; hence a lower apparent elastic modulus was derived from equation (3). Analysis of the bulk elastic modulus by the method of Wang et al⁵⁰ is therefore not considered reliable for Gilsocarbon graphite in this experimental setup. Valid measurements of the elastic modulus may be accomplished in this geometry by mapping the displacements on the surface with a higher precision than can be achieved by DIC, such as using ESPI (electronic speckle pattern interferometry)⁵². Recently, inverse modelling of DIC analysis of optical observations in flexure and disc compression (with a central hole) has been used to extract the engineering relationship between strain and stress in a fine grained isotropic graphite (IG11)^{72,73}. That work obtained a non-linear damage relation that was most significant in tension, with a decrease in elastic modulus with increasing strain that was also

significantly greater in tension than in compression⁷². The behaviour under biaxial loading was similar to uniaxial loading, with the non-linear damage effect slightly increased⁷³.

Figure 16: Effect of applied load (% of failure load) on a) the ratio of DIC-measured surface total strain magnitudes, at different temperatures, compared with the equivalent ratio for the total strains measured at room temperature by DVC of XCT images; the errors are smaller than the symbols; b) the development of DVC-measured hydrostatic strain at room temperature; c) the ratio of ND and XRD-measured elastic strain magnitudes at different temperatures up to 850°C.

In the disc compression geometry, for a continuum solid following a linear-elastic constitutive law with the same elastic properties in tension and compression, the compressive elastic strain at disc centre is, theoretically, approximately three times the magnitude of the orthogonal tensile elastic strain. The tensile stresses at the centre of the flattened disc compression specimen are not significantly affected by the Poisson's ratio⁷⁴. The measured ratios of the total strain magnitudes in the central region of the disc compression specimen are summarised in Figure 16a as a function of applied load, relative to the maximum load. Very similar data are obtained at room temperature whether the strains were measured by DIC at the specimen surface or by DVC in the bulk. The total strain ratio, $|\epsilon_{xx}^{DIC}|/|\epsilon_{yy}^{DIC}|$, was

~3 at low load and decreased to ~2 with increasing load. The DVC data, which were recorded only above 25% of the maximum load, show an almost constant strain ratio $|\varepsilon_{xx}^{DVC}|/|\varepsilon_{yy}^{DVC}|$ of ~2. In addition, the average hydrostatic strain was calculated by averaging the three DVC strains (ε_{xx}^{DVC} , ε_{yy}^{DVC} and ε_{zz}^{DVC}) in the central gauge region where the strains can be considered as small strains. The through thickness strains, ε_{zz}^{DVC} , were small (~15% of ε_{yy}^{DVC} and ~30% of ε_{xx}^{DVC}). The hydrostatic strain is dominated by the compressive strain in response to the compressive load, and shows a linear volume decrease with increasing load (Figure 16b). The high temperature DIC strain data generally have higher compressive/tensile strain ratios at low load and approach the lower bound of 2 gradually with increasing load (Figure 16a). This decrease in the total strain ratio is caused by a greater increase in the tensile ε_{yy}^{DVC} strains, relative to the compressive ε_{xx}^{DVC} strains, as the applied load increases. These effects would be consistent with the development of microcracking damage^{41,23,25} that would decrease the tensile elastic modulus⁷⁵ as the cracks open. However, such tensile cracks would not affect the compressive modulus. This is also evident in the relationships between the elastic strains and bulk strain shown in Figure 14. The greater scatter of the tensile strains in Figure 14, in comparison to the compressive strains, may be caused by heterogeneity of tensile cracking; this would cause more significant local variations in the stress carried by regions of microstructure that are subjected to the same tensile strains. The compression data show some deviation from linearity at high strains, which may indicate the development of compressive damage.

At room temperature, the XRD value for the elastic compression/tension strain ratio, $|\varepsilon_{xx}^{XRD}|/|\varepsilon_{yy}^{XRD}|$, is approximately constant at around 5 (Figure 16c). The individual ND data points are too scattered to calculate their ratio of compressive to tensile strain magnitudes, but it is possible to make a comparison by using the average values for $|\varepsilon_{xx}^{ND}|/|\varepsilon_{yy}^{ND}|$ within ranges of the applied load. This is presented also in Figure 16c, which shows the strain ratio remains low with increasing load at high temperature (i.e. 600°C and above), whereas it increases quite significantly to the order of 10 with increasing load at lower temperatures (i.e. 400°C and below). The room temperature ND and XRD data are consistent with each other. The increase to high values of elastic strain ratio occurs because the elastic tensile strains, ε_{yy}^{XRD} or ε_{yy}^{ND} tend not to increase with increasing load (Figure 14), unlike the compressive strains, ε_{xx}^{XRD} or ε_{xx}^{ND} . These measurements further demonstrate the inability of the graphite to

sustain a significant tensile stress at lower temperatures, whilst it has a greater ability to sustain tensile stress and achieved higher elastic strains at higher temperatures.

The literature value for the room temperature tensile strength of Gilsocarbon graphite is about 19-20 MPa. The tensile strengths measured at room temperature in the current experiment, by diametral compression, are lower (~17 MPa), but there is good consistency between the independent experiment setups for the same specimen geometry. The difference is not due to specimen size; there is evidence of a size effect in polygranular graphite^{76,29}, but the size effect is not strong in Gilsocarbon due to the high frequency of significant defects in its coarse microstructure⁷⁷. These intrinsic defects are of the order of 0.5 mm in size^{24,78} and their propagation causes tensile failure^{41,44,79,23}. Equation (1) assumes linear elastic properties that are the same in tension and compression. The results obtained in this work (e.g. Figure 16a) and by others^{20-22,72,73} clearly show that this is not the case. It was observed by Awaji and Sato⁴⁹ that a correction factor was required to obtain the uniaxial tensile strength from the tensile stress calculated at failure under diametral compression, and that this factor was sensitive to the ratio of tensile to compressive strength of the material. They proposed a mode I/mode II failure criterion to estimate this correction factor. There is evidence, also, that strain energy criteria can describe the multi-axial failure condition in polygranular graphite^{80,81}. The room temperature tensile strength obtained in this work is lower than the reported uniaxial tensile strength predominantly due to the biaxial compressive/tensile stress state at the centre of the disc geometry. Further modelling, using these data as inputs for the non-linear properties of graphite, may allow deeper insights into the correction factors needed to interpret tensile strengths from disc compression strength tests and other multi-axial stress states.

Nonetheless, the tensile strength, calculated as the maximum tensile stress from equation (1), increased with test temperature (Figure 5d). It is also clear that the maximum tensile stress that can be carried by the microstructure in response to applied tensile strain was significantly higher at elevated temperatures (>600°C) than at lower temperatures (Figure 14b). These observations agree with the increase in flexural strength of Gilsocarbon graphite reported by Liu et al⁴¹ at temperatures up to 1000°C. The tensile strength in Gilsocarbon is determined by the propagation of existing defects^{23,24,46}. Their dimensions would not be affected significantly by temperature, so the elevation of the high temperature tensile strength implies

an increase in defect tolerance, i.e. improved fracture toughness. There is consistent with previous work using *in situ* high resolution tomography by Liu et al ⁴¹ where an increased Mode I fracture toughness and a rising resistance curve at 1000°C was observed. A reduction in the micro-cracking that accompanied crack propagation was evident at 1000°C compared with ambient. Liu et al ⁴¹ correlated the reduction in micro-cracking, the increase in flexural strength and fracture toughness at 1000°C to the partial closure of nano- and meso-scale lenticular Mrozowski cracks and the relaxation of residual stresses associated with it. After the cooling process from >2,700 °C during manufacture, tensile stresses in graphite grains were ‘frozen-in’ at ambient temperatures from the high defect concentration in Gilsocarbon graphite. Upon heating, these residual stresses are relaxed and it has been proposed⁴¹ that this aids the closure of the anisotropic expansion cracks to reduce their surface energy, and subsequently increases the capability of the material to sustain higher strain and stress at grain level (i.e. $\Delta P'$ in Figure 15 is reduced such that Δ_m has a higher percentage of Δd).

In summary, this study has shown that the properties of Gilsocarbon graphite are influenced by the development of tensile microcracking within its microstructure. Elevated temperatures increase the resistance to microcracking, and we deduce that there is an increase in defect tolerance, caused by an increase in fracture toughness at elevated temperature, that increases the tensile strength. A quantitative analysis to directly link the changes at nano-/meso-scale to the macro-scale mechanical behaviour in Gilsocarbon graphite is still lacking. The role of microcracking and other intrinsic deformation mechanisms that allow graphite crystals to accommodate inelastic strains, such as basal slip and kink formation⁸² during high temperature deformation (tension/compression/bending) is also unclear, and the effect of temperature on these mechanisms might be related to that observed in other polygranular layered materials at high temperatures, such as MAX-phases^{83,84}. The combined effects of temperature and fast neutron irradiation on these mechanisms in nuclear graphites may be studied by using the techniques for *in situ* strain measurement by diffraction and image analysis that have been developed in this work. This will provide new insights into the mechanisms by which fast neutron irradiation affects the elastic modulus, strength and fracture toughness of nuclear graphite.

5 Conclusions

The polygranular synthetic nuclear graphite (Gilsocarbon, IM1-24) has been studied *in situ* by combined imaging and diffraction methods with X-rays at ambient temperature and neutrons at temperatures up to 850°C. This allowed direct measurement of the elastic strains in the graphite crystals (i.e. stress) and the total applied strain state as compressive and tensile strains develop simultaneously in the gauge volume at the centre of the disc compression specimens. It was found that the tensile strength of the graphite increases with temperature but there is no significant effect of temperature on the development of compressive strains. The relationship between the tensile elastic strains and the tensile total strains is temperature dependent. This behaviour is consistent with the occurrence of tensile microcracking at high applied load, and it is deduced that the resistance to tensile microcracking increases at high temperature, particularly at 600°C and above.

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